PHYSICS OF STRENGTH AND PLASTICITY

Features of Thermoplastic Deformations of Quasi-Anisotropic 2D Layers of Indium

M. D. Raranskyi, A. V. Oliinych-Lysiuk, I. G. Kurek, O. O. Tkach, R. Yu. Tashchuk, and O. V. Lysiuk

Yuriy Fedkovych Chernivtsi National University,
2 Kotsyubynsky Str.,
UA-58012 Chernivtsi, Ukraine

The changes in character of deformations in thin indium layers as a function of temperature and directions in crystals during rigid low-temperature thermomechanical cycling in the range of 2–300 K are investigated. As determined, the growth of anisotropy in thermal-elastic deformations brings about 'negative' dilations in some crystallographic directions under the stress $\sim\sigma_B$ in the range of 15–(80)100 K. It occurs at the process of transition from isotropic polycrystalline contacts to quasi-anisotropic 2D-nanocontacts in this metal. Within the framework of a dislocation model, the mechanisms of accumulation of such level of mechanical tension under conditions of limited space and minimal mobility of dislocations are analyzed in indium layers. The height values of Peierls barriers for kinks at dislocations and the coefficients of their transparency in indium are estimated as well. High probability of overcoming the barriers arises when kinks solitons penetrate them (tunnelling) in such crystallographic directions for which Poisson's ratios $\nu \sim 0.5$. These directions can serve as relaxation channels for destructive stresses accumulated in the layer.

Key words: indium 2D nanolayers, negative dilations, Peierls barriers, kink tunnelling.

У роботі досліджено зміну характеру деформації тонких шарів індію в залежності від температури і кристалографічних напрямів при жорсткому низькотемпературному термомеханічному циклуванні в інтервалі 2–

Corresponding author: Alla Vasylivna Oliinych-Lysiuk
E-mail: a_oliynich@ukr.net

300 К. Показано, що посилення анізотропії термопружних деформацій у цьому металі при переході від об'ємних ізотропних полікристалічних до квазіанізотропних 2D-наноконтактів призводить до появи в деяких кристалографічних напрямках при напруженнях ~σ_B «від'ємних» дилатацій в інтервалі 15–(80)100 К. Проаналізовано механізми накопичення в шарах індію механічних напружень такого рівня в умовах обмеженого простору і обмеженої рухливості дислокацій. В рамках дислокаційної моделі оцінено висоту бар'єрів Пайєрлса для перегинів на дислокаціях в індії та коефіцієнти їх прозорості. Виявлено високу ймовірність їх подолання шляхом проникнення (тунелювання) перегинів-солітонів через бар'єри у тих кристалографічних напрямах, для яких коефіцієнти Пуассона $\nu \sim 0{,}5$ і які можуть слугувати каналами релаксації руйнівних напружень, що накопичились у шарі.

Ключові слова: 2D наношари індію, «від'ємні» дилатації, бар'єри Пайєрлса, тунелювання перегинів.

1. INTRODUCTION

Unique physical and chemical properties of indium and its compounds along with continuous expansion of the fields of their practical application stimulate constant interest of practitioners and scientists [1–3]. Despite the fact that this metal has been known for a long time and is widely used in high-tech fields of science and technology, the information about the nature of its deformations during thermal cycling under low temperatures is topical both scientifically and in practical terms, remaining incompletely studied. Thus, under rigid low-temperature thermal cycling and conditions of abrupt temperature change (2–300 K), the issue of indium contacts reliability arises, causing failures in their work [4].

One of the reasons of these problems may be anisotropy growth in thermal-elastic deformations ε_{ij} during the transition from isotropic contacts in bulk samples of indium to quasi-anisotropic micro contacts in micro-particle layers (for example, as in flip-chips [4]) and especially to 2D-nanocontacts (as in gravity wave sensors [5]).

It is known that in a wide temperature range, monocrystalline indium is a non-axial auxetic—a material with anomalous deformation properties and negative values of Poisson's ratio in certain crystallographic directions [6]. These directions, under appropriate circumstances, can significantly influence the magnitude of the mechanical energy of the whole lattice and the energy values of defects in the crystalline structure at low-temperature thermal-cycling loading [7]. This effect can become critical in the process of miniaturization of contacts for indium (due to different distribution of deformation energy in the

layer and fundamentally different behaviour of defects, for example, dislocations of discrepancy in auxetic and non-auxetic directions in different crystallites [7], *etc.*).

The transition from micro-contacts to 2D nanolayers can bring about increase of the role of individual grains in the layer and essential growth of anisotropy of its mechanical properties, especially when the temperature and directions in crystallites change. The reason why it occurs is the significant difference in the values of Poisson's ratio and thermal expansion in indium depending on the temperature and crystallographic directions which can vary in the wide range from -0.7 to 0.9 and from $-45 \cdot 10^{-6} \, \text{deg}^{-1}$ to $85 \cdot 10^{-6} \, \text{deg}^{-1}$, respectively [8].

Therefore, this work, being a logical continuation of our previous research, shows the impact of temperature and directions in crystals made on the nature of thermomechanical deformation in thin indium layers at rigid thermomechanical cycling in the range of $2-300$ K. It allows to determine possible mechanisms of their deformation at cryogenic temperatures.

2. EXPERIMENTAL AND THEORETICAL DETAILS

The relation (1) is commonly used in the modern theory of elasticity to describe the deformations caused by the thermomechanical action [9]. To evaluate three-dimensional deformations of a linear isotropic solid with coefficients of thermal expansion α and Poisson's ratio ν, the normal components of the strain tensor ε_{ij} are written as

$$\begin{Bmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \end{Bmatrix} = \frac{1}{E} \begin{bmatrix} 1 & -\nu & -\nu \\ -\nu & 1 & -\nu \\ -\nu & -\nu & 1 \end{bmatrix} \begin{Bmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \end{Bmatrix} + \alpha T \begin{Bmatrix} 1 \\ 1 \\ 1 \end{Bmatrix}. \tag{1}$$

Taking into account, that thermal expansion does not cause displacement components.

Using the denotations for total dilatation

$$\sum \varepsilon_{ii} = \varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz} \tag{2}$$

and sums of normal stresses

$$\sum \sigma_{ii} = \sigma_{xx} + \sigma_{yy} + \sigma_{zz}. \tag{3}$$

The Eq. (1) can be rewritten as

$$\sum \varepsilon_{ii} = 3\alpha T + \frac{1 - 2\nu}{E} \sum \sigma_{ii}. \tag{4}$$

However, the values of thermal expansion change with the tempera-

ture variation in different directions in the indium crystal significant-ly, similar to the changes in the values of Poisson's ratio and Young's modulus [6]. Therefore, the estimation of deformations that occur dur-ing thermal cycling of polycrystalline isotropic indium layers requires the temperature-spatial dependences of these characteristics to be con-sidered under the conditions of their miniaturization. This correction becomes even more significant when reducing indium objects to na-nosize. For the layers of the material, the thickness of which is much smaller than their linear dimensions, the role of the stresses created by each crystallite increases. Such layers can be assumed as quasi-anisotropic ones. Not only temperature means much but also other fac-tors like direction in which the deformation occurs; the nature of the substrate that stimulates the growth direction of certain crystallites and their growing conditions taken together influence as well. The types and properties of defects that appear during the growth of layers and the process of their utilization are also significant.

That is, to estimate the deformations of fields that occur during thermal cycling, it is necessary to provide appropriate adjustments in relation (1).

Taking into account that all parameters of indium in Eq. (1) change with temperature anisotropically acquiring positive and negative val-ues, we used temperature dependences of experimentally determined coefficients for thermal expansion and elasticity modulus S_{ij} [10] in our calculations of thermal-elastic deformations. Now Eq. (1) can be rewritten in the form:

$$\begin{Bmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \end{Bmatrix} = \frac{1}{E(T)} \begin{bmatrix} 1 & -\nu_{ij}(T) & -\nu_{ij}(T) \\ -\nu_{ij}(T) & 1 & -\nu_{ij}(T) \\ -\nu_{ij}(T) & -\nu_{ij}(T) & 1 \end{bmatrix} \begin{Bmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \end{Bmatrix} + \alpha_{ij}(T) \begin{Bmatrix} 1 \\ 1 \\ 1 \end{Bmatrix}. \qquad (5)$$

In Eq. (5), the values of tensor components of Poisson's ratios $\nu_{ij}(T)$ are calculated by Eq. (6)

$$\nu_{ij} = -\frac{S_{ij}}{S_{ii}}. \qquad (6)$$

Young's modulus $E_i(T)$ for crystals of tetragonal syngony in an arbi-trary crystallographic direction is determined by taking into account tensor components of elasticity compliance moduli S_{ij} and directional cosines δ_{ij}:

$$E_i' = [(\delta_{11}^4 + \delta_{12}^4)S_{11} + 2\delta_{11}^2\delta_{12}^2 S_{12} + 2\delta_{13}^2(\delta_{11}^2 + \delta_{12}^2)S_{13} +$$
$$+ \delta_{13}^4 S_{33} + (\delta_{12}^2\delta_{13}^2 + \delta_{11}^2\delta_{13}^2)S_{44} + \delta_{11}^2\delta_{12}^2 S_{66}]^{-1}, \qquad (7)$$

where

$$\delta_{11} = \cos\varphi\cos\psi - \sin\varphi\cos\theta\sin\psi,$$
$$\delta_{12} = \sin\varphi\cos\psi + \cos\varphi\cos\theta\sin\psi,$$
$$\delta_{13} = -\sin\theta\sin\psi,$$
$$\delta_{21} = -\cos\varphi\sin\psi - \sin\varphi\cos\theta\cos\psi,$$
$$\delta_{22} = -\sin\varphi\sin\psi + \cos\varphi\cos\theta\cos\psi,$$
$$\delta_{23} = -\sin\theta\cos\psi,$$

(8)

among which values δ_{11}, δ_{12}, δ_{13} denote the direction of longitudinal extension/contraction and δ_{21}, δ_{22}, δ_{23}—the direction of transverse extension/contraction [7].

3. RESULTS AND DISCUSSION

We estimated the dilatation value for indium films when the deformation is in the *XOY* plane. The evaluation is made taking into account the fact that films with different deposition methods and on different substrates may have different predominant crystallographic directions of orientation and that significant stresses in such systems occur only in the plane of the layer (Fig. 1).

Figures 2–5 show the results of thermal-elastic deformations modeдling. They are obtained according to the correlation (5) taking into account the direction of growth textures. As we can see, the components of the strain tensor change differently in the direction $\langle 110 \rangle$ with temperature increase at mechanical stresses of 10^3 Pa. Components ε_{xx} and ε_{yy} increase in a monotonous way (Fig. 2, curves *1* and *2*, respectively), but when passing through the maximum at temperature 120 K, ε_{zz} decreases and at 230 K comes into the region of negative deformations (Fig. 2, *a*, curve *3*). 'Negative deformation' in this case may mean that the component of deformation determined by the Poisson's ratio begins to outweigh the component caused by the coefficients of thermal expansion. It occurs because these coefficients remain positive up to temperatures ~280 K and the direction $\langle 110 \rangle$ becomes auxetic [6].

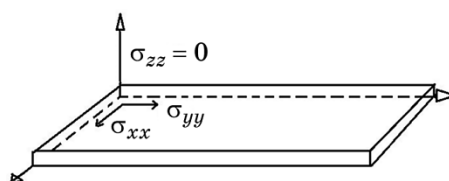The increase of mechanical stresses up to 10^8 Pa in the same crystal-

Fig. 1. Schematic representation of stress distribution in a 2D indium layer.
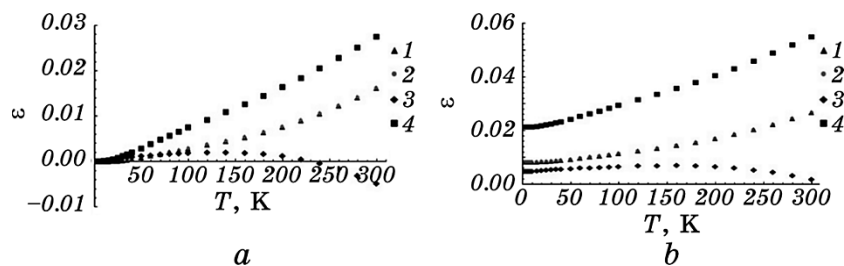
Fig. 2. Temperature dependences of strain tensor components ε_{xx}, ε_{yy}, ε_{zz} (curves *1–3*) and the values of total normal deformation ε_{ij} (curve *4*) for 2D indium layers, given in *XOY* plane with direction $\langle 110 \rangle$ under conditions of external compressive strain 10^3 Pa (*a*) and 10^8 Pa (*b*).

lographic direction does not change the behaviour of the components ε_{xx} and ε_{yy} with temperature increase (Fig. 2, *b*, curves *1* and *2*, respectively). The maximum value for the component ε_{zz} shifts to the right on the temperature scale (up to 150–160 K) and acquires negative values at the temperature above 300 K (Fig. 2, *b*, curve *3*). In this case, the total deformation values increase from 0.02 to 0.056 remaining positive throughout the whole range of thermal cycling temperatures (Fig. 2, *b*, curve *4*).

The picture similar to that described in Fig. 2, *a* is also observed in direction $\langle 001 \rangle$ (Fig. 3, *a*). However, as soon as the value of σ increased up to 10^8 Pa—values ε_{zz} became negative in the whole temperature range (Fig. 3, *b*, curve *3*). It changed the sign of the total thermal elastic deformations parameters to the opposite in temperature interval from 0 to 100 K. Thus, the temperature dependences of the deformation of quasi-anisotropic indium layers in different crystallographic directions practically do not change at stresses 10^3–10^6 Pa. But, when reaching stresses of magnitude 10^8 Pa, they become fundamentally different: we observed gradual increase in directions $\langle 110 \rangle$, $\langle 101 \rangle$, $\langle 100 \rangle$ with positive values, and the inversion takes place in direction $\langle 001 \rangle$ at 80–100 K (Fig. 3).
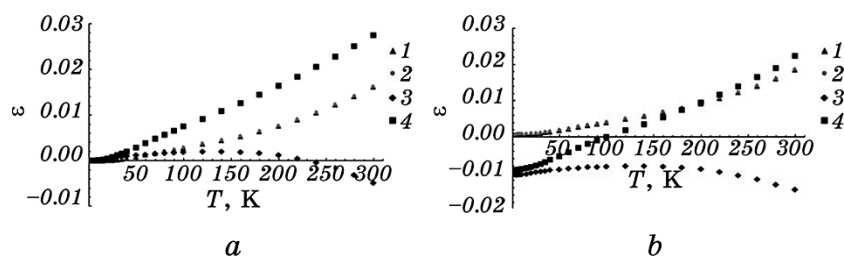

Fig. 3. Analogous dependences of strain tensor components of indium in direction $\langle 001 \rangle$ at external compressive deformations 10^3 Pa (*a*) and 10^8 Pa (*b*).

It is known [11, 12] that polycrystalline indium at room temperature has low strength properties and high level of resilience (elasticity) but when decreasing the temperature to 77 K its strength characteristics slightly increase: for example, σ_B changes from $2.94 \cdot 10^6$ Pa at room temperature reaching $1.275 \cdot 10^7$ Pa at 170 K and it remains the same up to 77 K. That is, when the temperature rises above 170 K, so-called failure of strength takes place—its value decreases more than twice. However, only relative values of stretching for the specimens (tensile deformation) demonstrate changes almost 1.5 times as opposed to the relative squeezing (compression deformation) which values decrease only to 16% (see Table 1) [11].

The estimation of the degree of compression deformation caused by thermal stresses $\varepsilon_T = 3\alpha T$ showed that, for indium at room temperature, it would be of the order of 0.045 in direction $\langle 001 \rangle$ and -0.0225 in directions $\langle 100 \rangle$ and $\langle 010 \rangle$, respectively. It is obvious that such deformations are not purely elastic and they will be realized not only due to atoms displacement, but this also can be caused by motion and interaction of defects, including linear ones. However, at ~77 K the thermal stresses will differ neither in the magnitude nor sign (positive or negative), since the values of the thermal expansion coefficients at this temperature are equal to each other $\alpha_\perp = \alpha_\parallel$ [8]. Lowering the temperature below 77 K will lead to the thermal stresses increase, but it will exclude the movement of some defects from the deformation process, in particular dislocations. It will accumulate stress in the layer magnifying it to the order σ_B. It is clear that the magnitude and sign of the total deformation at such stresses σ_B will vary depending on the direction in the crystal and the value of mechanical stresses, especially at temperatures below 170 K. This is confirmed by the results represented in Figs. 4, 5, where the temperature dependences of the total normal deformation are reproduced in the quasi-anisotropic layers of indium under compression (Fig. 4, a) and stretching (Fig. 4, b) with σ values varied from 10^6 to 10^8 Pa.

As we can see, in the so-called 'auxetic' directions (in bulk samples of monocrystalline indium), the compression deformations increase

TABLE 1. Effect of temperature change for the mechanical properties of highly pure (99.999%) indium measured at strain deformation rates $\dot{\varepsilon} = 2.5 \cdot 10^{-2} \, \text{s}^{-1}$.

T_{test}, K	σ_B, MPa	δ, %	ψ, %
77	12.75	35	92
172	12.75	34	82
215	5.88	38	96
258	3.94	50	98
301	2.94	57	98

with increasing magnitudes of stresses and temperature remaining exclusively positive (Fig. 4, *a*), and they decrease at stretching gradually moving into the area of negative values. At the same time, we observe the mirror-like situation while compressing in direction $\langle 001 \rangle$: the deformation decreases with the increase of temperature and mechanical stress and at the values of $\sigma \sim 10^8$ Pa it becomes negative. When stretching, the deformation increases and remains with positive values throughout the whole temperature and stresses ranges.

Thus, stretching indium crystals in the directions $\langle 110 \rangle$, $\langle 101 \rangle$, $\langle 100 \rangle$ at stresses of hundreds of MPa leads to the decrease of the amount of deformation with the increase of mechanical stresses and to the inversion of the deformation sign in the range 0–80 K. It is not a surprising fact when taking into account that the directions $\langle 110 \rangle$ and $\langle 101 \rangle$ in bulky material are auxetic, *i.e.*, the deformations in these directions are anomalous and the crystals should swell when stretching. But in the direction $\langle 001 \rangle$ the situation is quite opposite to the one discussed above: the reduction of deformation and its inversion are observed in the interval 0–100 K during compression, though this direction is not considered to be auxetic in bulk samples of indium but, when stretching, the deformations values remain with positive signs (Fig. 5, *b*).

To find out what lies in the basis of the results obtained, let us consider the process of layers deformation at cryogenic temperatures under the conditions of limited space and motion for linear defects. It is known that the residual change in the shape of real crystals occurs due to the movement of dislocations and at low temperatures (of the order of several tens of K) the movement of dislocations can be realized mainly by means of displacement of geometric kinks [13]. In fact, the dislocations movement is limited in this temperature range by energy barriers known as Peierls barriers. The energy value of such barrier E_P can be estimated using the relation:
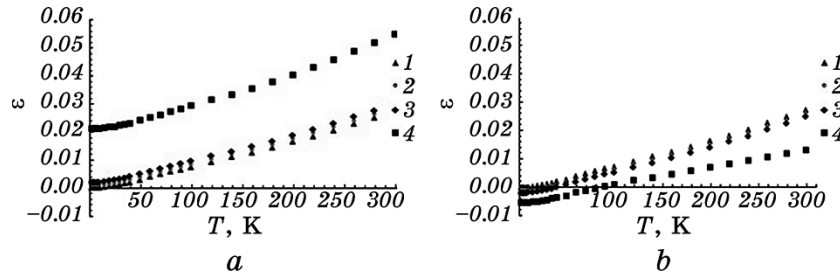

Fig. 4. Temperature-strength dependences of the total normal deformation $\sum \varepsilon_{ij}$ for 2D indium layers in *XOY* plane and in direction $\langle 110 \rangle$ with different external deformations of compression (*a*) and stretching (*b*): *1*—10^6 Pa, *2*—$0.5 \cdot 10^7$ Pa, *3*—10^7 Pa, *4*—10^8 Pa.

$$E_P = \frac{Gb^3}{300d},$$

where G is the shear modulus, b is the Burgers vector, d—the inter-planar distance [14]. The opportunity to overcome these barriers for edge dislocations $E_p \sim 1.2 \cdot 10^{-21}$ J at low temperatures in indium is almost impossible. However, the kinks formed at the dislocations during both the deformation process and at the dislocation origination when cultivating the layers, can overcome the barrier due to the phenomenon of tunnelling [14]. It is known, that the process of moving kinks 'under barrier' can be considered as solitons tunnelling, since the kinks at dislocations are associated with these separated nonlinear waves which interact with potential characteristics of the lattice [14]. As it follows from quantum mechanics, the tunnelling effect will be significant if the correlation takes place:

$$\exp\left(-\frac{2l}{\hbar}\sqrt{2m_s(E_P - E_K)}\right) = 1, \qquad (9)$$

where $l = l_{\text{In}} \sim 10^{-10}$ m —the width of the kink, $\hbar$—reduced Planck's constant, $m_s = (0.2 - 0.5)m_p = 8.36 \cdot 10^{-28}$ kg —the mass of the kink-soliton (assuming that the mass of the soliton is half the mass of the proton) [14, 15]. The estimation of the kinetic energy of the kink E_K carried out according to the equations represented in [13] showed that the kinetic energy of soliton in indium $E_{K\text{in}}$ can be $\sim 1.02 \cdot 10^{-29}$ J. Hence, the tunnelling coefficient is

$$\exp\left(-\frac{2l_{\text{In}}}{\hbar}\sqrt{2m_s(E_{P\text{In}} - E_{K\text{In}})}\right) = 0.065.$$

That proves the fact of high probability to overcome Peierls barriers by the kinks at dislocations in indium. This remarkable result indicates that the quantum tunnelling phenomenon is inherent not only for monoatomic systems (such as He atoms, for example) but also for ordi-
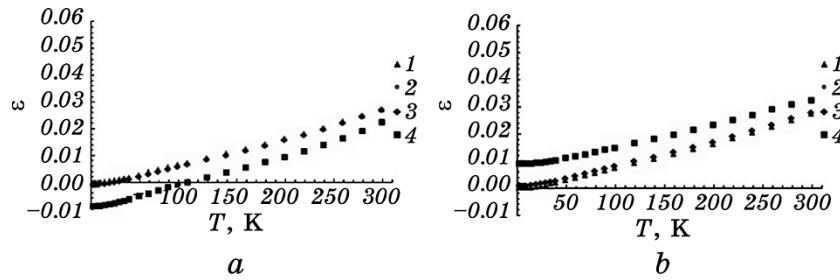

Fig. 5. The same dependences given in the direction of $\langle 001 \rangle$ (ν_{31}).

nary crystalline systems where atoms do not exhibit quantum properties under these conditions where as their defects possess such properties [14, 15]. The probability of quantum tunnelling for atoms can be estimated using the de Boer parameter Λ [15]:

$$\Lambda \sim \hbar(mu)^{-1/2} / a, \tag{10}$$

where m is the mass of the atom (soliton), u is the energy of interaction for adjacent atoms (solitons) which are at distance a. Suppose that the soliton energy is determined by the energy of the elastic interaction of double kinks at dislocations in the crystal, then this kind of energy can be found using the relation (11), (see [13], [15]):

$$u = \frac{Gb^2a^2}{8\pi L} \frac{(1 - 2\nu)}{(1 - \nu)}, \tag{11}$$

where L—the distance between kinks.

From the calculations above, it is clear that binding energy u between solitons (and, hence, de Boer parameter) depends on the magnitude and the sign of the Poisson's coefficients which are different in different crystallographic directions of indium. Fig. 6, a represents the results of values u estimated for the whole range of changes of Poisson's coefficients in indium (varied from 0.9 to −0.7). The function $u(\nu)$ undergoes inversion when the coefficient $\nu = 0.5$. The estimation of the de Boer parameters showed that kinks-solitons have a relatively high probability of tunnelling (0.15−0.3) at dislocations in indium and this opportunity increases when approaching $\nu \sim 0.5$ (Fig. 6, b). Thus, the deformation in indium at low temperatures can take place due to tunnelling dislocation kinks in the directions for which Poisson's coefficients $\nu \geq 0.5$, creating specific channels of super-elasticity along which accumulated stresses relax. That is, such channels will
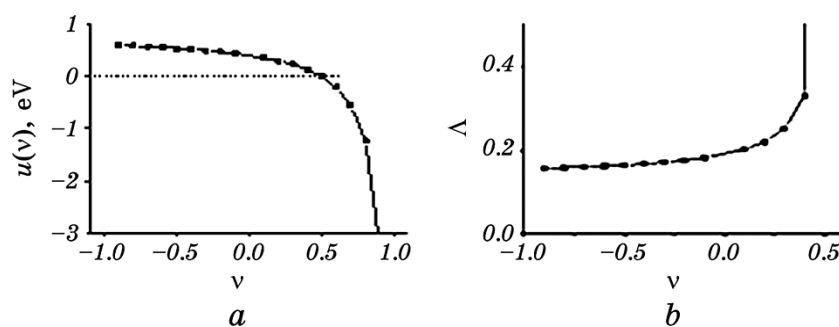

Fig. 6. Dependence of the binding energy of the dislocation kinks (*a*) and de Boer parameters (*b*) on the magnitude and signs (positive or negative) of the Poisson's coefficients.

emerge with high probability only when textures of growth in the process of indium layers cultivation are formed in such crystallographic directions where $\nu_{12} \geq 0.5$ and $\nu_{13} \sim 0.5$.

This assumption is confirmed to some extent by the change in the character of the displacement fields around the edge dislocations in indium (Fig. 7), when the coefficient changes from -0.5 to 0, the field around dislocation with the growth of the value ν changes slightly but when $\nu \geq 0.5$, it becomes fundamentally different.

It is interesting to note that it is in the range of $15-80$ K in the indium nano-particles with a diameter of $7-8$ nm that the authors [16] observed tunnel electrical conductivity. It is the evidence of overlap wave functions of the electrons in nano-particles, which allowed them to overcome the potential barrier of thin dielectric layers of glass by means of penetrating the barrier. With temperatures increase above 100 K, the tunnel conductivity in nano-particles became diminishingly small. Note also, that at temperature below 15 K indium demonstrates
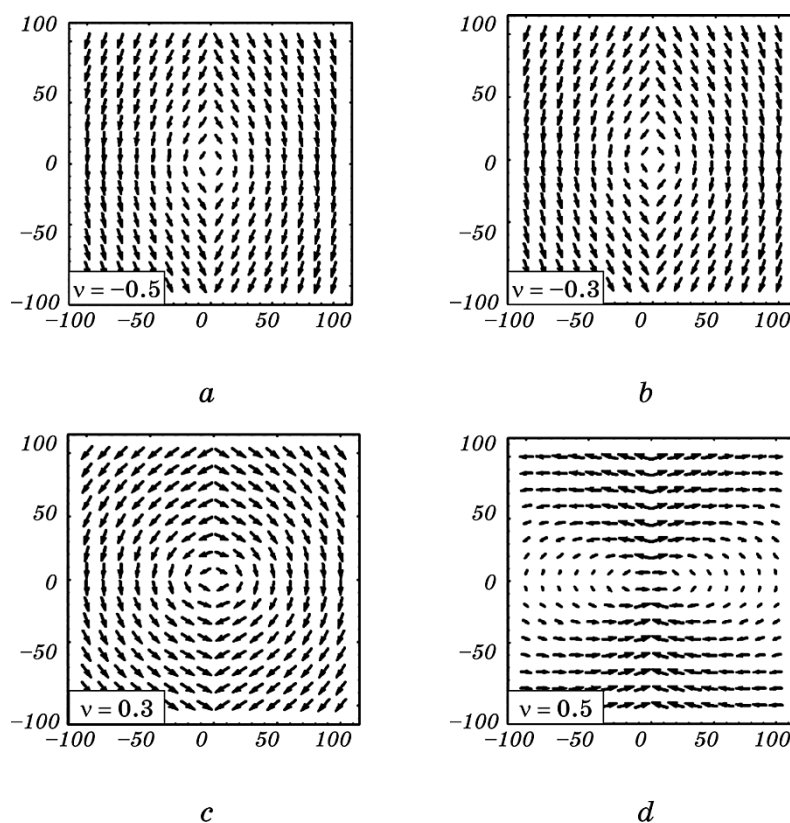

Fig. 7. The scheme of stress fields distribution around the edge dislocation in indium at different values of the coefficient ν.

a number of anomalies for thermal expansion coefficients associated with the ability of this material to transfer to the state of superconductivity [8]. Thus, with data of temperature-spatial dependences of ν_{ij}, $E_i(T)$ and $\alpha_{ij}(T)$ for bulk indium in a wide temperature range, it is possible to identify the deformation characteristics in each crystallographic directions for 2D indium layers when modelling their thermomechanical properties. It allows to predict the possible relaxation mechanisms of mechanical stresses in such layers taking into account their cultivation technology and conditions of thermal cycling.

4. CONCLUSION

1. The temperature dependences of thermomechanical deformation in 2D layers of indium in temperature interval 0−300 K have been modelled and analyzed. It is shown that compression deformations occurring in direction $\langle 001 \rangle$ will be of negative value at the temperature up to 100 K under rigid thermal cycling. Tensile deformations in directions $\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 101 \rangle$ will be negative at temperatures up to 80 K.
2. The coefficients transparency of the Peierls barriers and de Boer parameter for kinks at dislocations in indium are evaluated. It is revealed that with wide range of Poisson's coefficients there exists a rather high probability of kinks tunnelling at dislocations (0.15−0.3) reaching their maximum (≥0.5) at $\nu \sim 0.5$.
3. A mechanism of relaxation for mechanical stresses in indium layers under conditions of limited mobility of dislocations in super-elastic channels is proposed. Those channels emerge in the directions where $\nu \sim 0.5$. It is indirectly confirmed by the behaviour of the vector stress fields around boundary dislocations and by the presence of unique sealing properties in indium alloys.

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